PARTITIONING AND TRANSMUTATION OF NUCLEAR WASTE: THE DUTCH RAS PROGRAMME AND ITS RELATION TO INTERNATIONAL STUDIES

R.J.M. Konings, H. Gruppelaar, E.H.P. Cordfunke and J. Prij
Netherlands Energy Research Foundation ECN,
P.O. Box 1, 1755 ZG Petten, Netherlands

ABSTRACT

In the present paper a review of the Dutch RAS programme is presented. The objectives and achievements of the programme are described. Special attention is given to a recent assessment of the international status of P&T that has been made for the Dutch authorities. The major conclusion of this assessment is that P&T is an important instrument in the management of nuclear waste to achieve the technical limits (ALARA) with respect to radiotoxicity of the waste inventory as well as radiological effects of the disposal. The technology for P&T is, however, only partially available at present and recommendations for the directions of the RAS programme are given.

1. INTRODUCTION

Spent fuel elements from nuclear power plants contain many radioactive elements (actinides, fission products) which are a possible thread to mankind if released to the biosphere. Disposal in geological repositories is considered to be the final destination of spent fuel or the waste produced by reprocessing of the fuel elements, but is, however, not considered safe by a significant fraction of the population in many countries. This could be overcome if the actinides can be removed efficiently from the spent fuel and the radiotoxicity of the waste can be reduced to that of the uranium ore it was produced from, in a reasonable period of time (e.g. 250 years). Reprocessing, partitioning, recycling and transmutation are techniques to achieve this goal.

In 1991 the Netherlands Energy Research Foundation ECN started a research programme on recycling and transmutation of long-lived nuclides that are present in the spent fuel from nuclear power generation. This programme, which is known under the Dutch acronym RAS, is concentrated on the following five topics:

- reactor physics and scenario studies,
- chemical and material studies of fuels and targets,
- irradiations in the High Flux Reactor at Petten,
- radiological effects and risks,
- non-proliferation.

In the present paper a short description of the objectives and achievements of the RAS programme is given, followed by the present views on the state-of-the-art of partitioning and transmutation of actinides and fission products. Strategies and (innovative) fuel cycle technology required for the recycling of plutonium, minor actinides and fission products are discussed and their possibilities and limits are identified. Also the potential of future options with low actinide production (thorium cyclus, accelerators) is considered. An extended report on this work is given in Ref. [1].

2. OBJECTIVES AND ACHIEVEMENTS OF THE RAS PROJECT

In the Dutch RAS programme the ALARA principle is applied to the radiotoxicity of the waste as well as to the risk of disposal to the benefit of future generations. As a consequence, the investigations in the field of P&T are focused on:

1. the actinides, in particular Pu and Am, which dominate the medium- and long-term radiotoxicity of HLW,
2. long-lived mobile components (in particular Np, Tc, I, Cs), which dominate the long-term radiological effects of disposal.

However, the justification of the work is not based on scientific arguments since the risks of geologically stored waste to future generations are estimated to be very low. The RAS programme should rather demonstrate the maximum possible measures that are necessary to relief public concern.

In the first phase of the RAS programme considerable attention was given to the transmutation of the fission products technetium and iodine. In the frame of the EFTTRA cooperation [2], an irradiation experiment (EFTTRA-T1) was performed in the HFR to study the behaviour of several target materials [3-5]. In parallel, scenario studies for the transmutation of these fission products in various reactor types have been made [6,7]. The overall conclusion of the work is that transmutation of technetium is technically possible although the irradiation times in most reactor types are (very) long and the technetium inventories are high. The transmutation of iodine in existing fission reactors does not look feasible (see below).

Currently, the RAS programme is focused on the transmutation of actinides. Different recycling modes for plutonium and americium are being studied by calculational methods and experiments, including MOX fuel in current LWRs and TRU fuels in future reactors. Again, the efforts within the EFTTRA programme are an essential part of this work: three irradiation experiments are planned in the HFR (Table 1) to study the irradiation behaviour of various ceramic materials that can be used as inert matrix in non-fertile fuels or targets [8]. In addition, laboratory experiments are performed to characterize a number of inert-matrix materials and to determine their physico-chemical properties. Various scenarios for the transmutation of plutonium and americium have been analysed [9-11]. An overall conclusion is not yet available but the preliminary results show that recycling of Pu is possible, whereas recycling of Am is only feasible if the technological problems with respect to reprocessing and fuel fabrication can be solved, as will be discussed below.

The use of the thorium cycle in fission reactors as a means to minimize the production of actinides has also been investigated. The state-of-the-art of the technology of thorium-based fuel has been reviewed [12] with respect to reactor physics, chemical and material aspects and actinide waste. In this study it was concluded that a substantial reduction of actinide radiotoxicity can be achieved by using thorium-based fuels if efficient partitioning and multiple recycling of uranium and thorium can be realised. Finally, a limited effort is devoted to the study of accelerator-based transmutation. including contributions to nuclear data for these systems [13].

In 1996, an assessment of the international status of P&T has been made for the Dutch authorities in order to define the next phase of the RAS programme [1]. The report has been reviewed by a national committee of experts from government, science and industry. In the following three sections, the views expressed in this report are summarized.

Table 1. Irradiation programme in the HFR at Petten.

Experiment	Description	Period
EFTTRA-T1	transmutation of the fission products technetium (~6% burn-up) and iodine	1994
EFTTRA-T2	• re-irradiation of technetium (> 20% burn-up) • irradiation of ceramic materials for the development of fuels for actinide burning (inert matrices)	1996-1998
EFTTRA-T3	irradiation of inert matrices, mixed with enriched uranium	1996-1997
EFTTRA-T4	demonstration of the transmutation of americium in an inert matrix	1996-1999

3. STATUS OF PARTITIONING AND TRANSMUTATION

3.1 Plutonium and Uranium

Separation of plutonium (Pu) and uranium (U) from spent fuel will lead to a decrease of the radiotoxicity of the remaining waste by a factor 10. Using current PUREX technology, Pu and U can be separated from spent fuel with high efficiency (99.5-99.88 %). From the point of view of P&T, further improvement of the separation efficiency is only useful if also the minor actinides, in particular americium, are removed from the high level waste.

Pu and U can be recycled in thermal reactors in the form of mixed oxide (MOX) fuel. In case of 30 % MOX loading, as is current practice, Pu recycling in LWRs will slow down the growth of plutonium stocks. Higher MOX loadings (up to 100 %) will lead to net Pu consumption in future reactor designs. The effect of Pu recycling in LWRs on the radiotoxicity is, however, limited due to build-up of non-fissile Pu isotopes. Ultimately, the use of reactors/devices with fast neutron spectra is inevitable to reach substantial reduction of the plutonium radiotoxicity. The technology for the design of devices and fuels with maximum incineration rates needs to be developed.

3.2 Minor Actinides

Partitioning and transmutation of minor actinides, and especially americium, is necessary because a reduction of the radiotoxicity greater than 10 is wanted. If separation is realised with a 99-99.9% efficiency, a reduction of the radiotoxicity by a factor greater than 100 is within reach, provided that recycling is performed efficient as well. However, at present partitioning of the minor actinides americium and curium from PUREX waste solutions is not yet possible at an industrial scale. In several countries liquid-liquid extraction processes have been developed (Table 2) but in all cases the lanthanides, an important group of fission products, are co-extracted as a result of their chemical similarity to the trivalent actinides. New techniques to separate the trivalent actinides and lanthanides as well as the actinides mutually are required. The use of macrocyclic extraction molecules in Supported Liquid Membranes (SLM) is a promising development in this field and is studied in several European laboratories, among which ECN.

Transmutation of the minor actinides can be done efficiently in a fast-neutron flux but also the benefits of a "once-through" incineration in a thermal flux have to be evaluated. Neptunium can be recycled in MINOX fuels, americium in special targets containing an inert matrix. In practice, recycling of curium does not seem feasible at the moment because of the α-, γ- and neutron doses due to decay and spontaneous fission which extremely complicate the fabrication curium fuels and targets.

When transmutation of the minor actinides is introduced in the fuel cycle, existing non-proliferation measures have to be extended to cover P&T. Most of the problems to the minor actinides are similar to those of reprocessing and MOX fabrication plants which have been proven to be adequately safeguardable. However,

Table 2. Liquid-liquid extraction processes for the trivalent actinides.

process name	country	extraction molecule	HNO_3 concentration	typical results
DIAMEX	France	diamides	0.5-5 M	Am+Cm > 95%
DIDPA	Japan	DIDPA/TBP	0.5 M	Am+Cm > 99.9%
TRPO	China	TRPO	0.1 - 1 M	Am+Cm > 95%
TRUEX	USA	CMPO/TBP	0.1 - 3 M	Am+Cm > 99.9%

specific P&T issues need to be addressed, such as the potential of mis-use of minor actinides and the new facilities. Until today the civil fuel cycle has shown to be a rather difficult route for proliferation and it is of utmost importance to maintain this characteristic also for new installations and fuel cycles.

3.3 Fission Products

Partitioning and transmutation of long-lived fission products is only relevant from the point of view of reduction of radiological effects. In this respect, the following three fission products need to be considered: cesium (^{135}Cs), iodine (^{129}I) and technetium (^{99}Tc). The technology to separate these elements from HLLW is available on a laboratory scale, but has not been implemented in the PUREX process. Technetium and iodine can be transmuted by single neutron capture, but the transmutation half-lives and inventories in most reactor types are high. In addition, transmutation of these fission products requires additional enrichement. Transmutation of cesium is not feasible because of the low neutron absorption cross section of ^{135}Cs and parasitic absorptions in ^{137}Cs and ^{133}Cs which are present in the fuel in about equal quantities as ^{135}Cs.

3.4 Survey of transmutation devices

Recycling of plutonium is possible in existing LWRs with respect to reduction of masses. However, as multiple recycling of Pu in LWRs is limited and transmutation of minor actinides in existing thermal reactors is not effective, a significant radiotoxicity reduction is only obtained in follow-up transmutation in dedicated burners. These burners should have low Pu production, a high specific power and for minor actinide incineration, a fast neutron spectrum. Nearby technology indicates CANDU and CAPRA burner types as interesting candidates; future developments hint in the direction of accelerator-based systems. Also fission-product transmutation can best be achieved in dedicated thermal burners or in moderated subassemblies of fast reactors since transmutation half-lives and inventories in LWRs are very high. The potential of existing, near-by and future reactor technology is summarized in Table 3.

Table 3. Review of the recycling capabilities of various transmutation devices; MA denotes minor actinides, FP fission products.

device	Pu	MA	FP
LWR	limited[a]	poor	no
HWR	moderate[a,b]	poor	moderate
HTR	limited[c]	poor	no
ALMR	good	good	no
CAPRA	excellent	good	reasonable[d]
Pb-cooled burner	excellent	good	reasonable[d]
molten salt reactor	good?	?	?
accelerator-driven thermal burner	good?	?	good
accelerator-driven fast burner	excellent	good	?

[a] Multirecycling of MOX is limited due to production of even-mass Pu and other actinides.
[b] 100% Pu in an inert matrix.
[c] Reprocessing is difficult.
[d] In a moderated subassembly.

4. OPTIONS WITH LOW ACTINIDE PRODUCTION

4.1 Radiological clean energy production?

The use of pure fissile material, in particular fissile U isotopes, gives lowest long-lived radiotoxic actinide production. This requires fissile material from either (high) enrichment or breeding. Use of uranium, highly enriched in ^{235}U (HEU), is not recommended from the point of view of non-proliferation. The alternative is the use of ^{233}U produced in the thorium cycle, which is more proliferation resistant, due to hard γ-rays from co-generated ^{232}U. At present, extension of burnup of LWR fuel is a good measure to limit the actinide production per unit of generated electricity in the current open U/Pu cycle.

4.2 Thorium cycle in reactors

The effective use of thorium in an open cycle requires fissile topping material and a high burnup. The radiotoxicity of waste is lowest if ^{233}U or ^{235}U (HEU) is used as topping material. Further reduction of the radiotoxicity is obtained in a closed cycle using thermal reactors and ^{233}U or ^{235}U (HEU) as topping material and recycling of uranium. The best result should be obtained in a fast reactor with recycling of all actinides and without topping material. The main obstacle for introduction of the closed cycle is that reprocessing and fuel fabrication have to be introduced on an industrial scale, including adequate safeguards. A good introduction to the thorium cycle could be to burn Pu in a thorium oxide fuel matrix.

4.3 Accelerator-based energy production

Accelerator-based systems have probably some safety advantages and produce excess neutrons. These two aspects give flexibility to design future systems for safe, clean and acceptable nuclear energy production and/or waste transmutation. The accelerator offers the possibility for applying a closed thorium cycle, due to excess neutrons (when coupled to thermal reactors) or additional safety (when coupled to fast reactors). The Fast Energy Amplifier is one of the examples with high potential. Other advanced features are related to waste transmutation of difficult long-lived components, like minor actinides and long-lived fission products and the possibility of incinerating the radioactive inventory at the end of a nuclear era.

5. CONSEQUENCES FOR DISPOSAL

The radiological effects of disposal of waste in a (geological) repository are not only determined by the radiotoxicity but also by the performance of the natural and engineered barriers of the disposal concept. The safety-relevant radionuclides that are identified 'normal evolution' release scenarios used in performance assessments, are the fission products ^{135}Cs, ^{129}I, ^{79}Se, ^{99}Tc and the actinide ^{237}Np and its decay products. However, the calculated maximum exposures in 'altered evolution' scenarios are far below the natural background. For human intrusion scenarios the maximum exposure is more significant and in these scenarios the actinides ^{241}Am, ^{243}Am and ^{240}Pu predominate the radiological effects.

Table 4. Some results of performance assessments for disposal of vitrified HLW in a salt formation in the Netherlands.

Scenario	Maximum exposure		Dominant radionuclides
	magnitude (Sv/y)	time (y)	
Normal evolution	10^{-7}	10^{6}	^{237}Np, ^{233}U, ^{135}Cs
Ground water intrusion	10^{-8}	10^{6}	^{135}Cs, ^{79}Se, ^{226}Ra
Human intrusion	10^{-4}	500	^{241}Am, ^{243}Am, ^{240}Pu

It should be realised that the separation of the actinides alone, as a result of which the radiotoxicity of the waste is reduced below the level of natural uranium ore after 250 years, does not imply that disposal in a (geological) repository is no longer needed. But if such waste is disposed in a carefully selected geological formation, the risk can be as low as that of natural uranium ore in its geological situation.

6 CONCLUSIONS

Partitioning and transmutation of actinides are important elements in the management of the waste from nuclear power generation. They are means to achieve the limits (ALARA) with respect the radiotoxicity of the inventory of disposal sites as well as the radiological effects of human intrusions of disposal sites in the future. The technology required for the implementation of P&T of actinides in the fuel cycle is only partially available at present and much research is still needed. In the coming years the efforts of the RAS programma should be concentrated on:

- Improved partitioning methods for trivalent actinides.
- Transmutation of actinides using non-fertile fuels.
- Scenario studies using 100 % MOX, HWRs and fast burners.
- Innovative systems for future "clean" energy production and transmutation using the thorium cycle and/or accelerators.

To reduce the (small) effects of exposure of radionuclides due to normal evolution, transmutation of long-lived fission products should also be realised. However, on the basis of the present technology this does not seem feasible and partitioning and immobilisation might be a more realistic way of achieving this. Study of this topic is recommended.

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